FROM:

08/665,343 filed June 17, 1996 which is a Continuation-in-part of U.S. Serial No: 612,586 filed March 8, 1996; PCT/US94/04278 filed 4/19/94 (published 5/26/95 No. WO95/13851); PCT/US94/07314 filed 6/27/94 (published 1/4/96 No. WO 96/00118); 288,690 filed 8/11/94; 581,188 filed 12/29/95; 581,191 filed 12/29/95; 581,125 filed 12/29/95. In turn U.S. Serial Nos. 581,188; 581,191; and 581,125 are continuation-in-parts of the following applications: Serial Nos.: 288,690, filed August 11, 1994, PCT/US94/07314 filed June 27, 1994 (CIP of PCT/US 94/04278, filed 19 April 1994)."

and insert:

--- 8/8/98; 08/984,459, filed 12/3/97; 08/909,487, filed 7/12/97; 08/863,794, filed 5/27/97; 08/819,675, filed 3/17/97 now Pat. No. 5884639; PCT/US97/17534, filed 9/30/97 now Ser. No. 09/230,940 filed 2/3/99; Ser. No: 08/719,817, filed-9/30/96; Ser. No: 08/665,343, filed 6/17/96 which is a CIP Ser. No: 08/612,586, filed 3/8/96; PCT/US94/04278, filed 4/19/94 now Ser. No. 08/211,781 filed 5/14/96, Pat. No. 6,033,283; PCT/US94/07314, filed 6/27/94 now Ser. No. 08/256,235 filed 6/27/94 and Pat. No. 5,868,597; 08/288,690, filed 8/11/94 now Pat. No. 5,633,286; 08/581,188, filed 12/29/95 now abandoned; 08/581,191, filed 12/29/95 now Pat. No. 5,760,117; and 08/581,125, filed 12/29/95 now Pat. No. 5,962,572. In turn Ser. Nos./581,188; /881,191; and /581,125 are CIPs of the following applications: Ser. Nos.: 08/288,690 and PCT/US94/07314 which in turn is a CIP of PCT/US 94/04278, filed 4/19/94. ---

At page 2, line 32 and line 37; at page 4, lines 11 and 16; at page 7, lines 7, 8, and 37; at page 8, —lines 14, and 16; at page 9, lines 14, 15, 17-20, and 22; at page 10, line 31; at page 11, lines 5, and 7; at page 17, lines 25, 27, 29, 31, 33, 36, and 37; at page 18, lines 2, 4, 5, 7, 11, and 13; at page 19, line 6; at page 20, lines 9-13; at page 24, lines 21-22; at page 25, lines 19, 21, 25, and 26; at page 26, lines 21-23; the degree notation "o" should be correct to read the degree as a superscript — 0 — respectively.

5,633,286

At page 9, line 27, correct "CH2" to read --- CH<sub>2</sub> ---.

At page 9, line 34, correct "(-CH2-)16" and "(0.67)4" to read --- (-CH<sub>2</sub>-)<sub>16</sub> --- and --- (0.67)4 --- respectively.

At page 9, line 37 and top of page 10 line 1, correct "(0.67)4" and "(-CH2-)4" to read --- (0.67)4 ---  $^{\circ}$  and --- (-CH<sub>2</sub>-)<sub>4</sub> ---.

At page 10, line 1-8 delete: "which is the percentage of (0.67)4 or "(-CH2-)16" units. Thus, when n O = 28 or 72% of (-CH2-)4 units, the % crystallinity is (0.72)4 or 26.87% crystallinity attributed to (-CH2-)<sub>16</sub> units, denoted by -EB<sub>28</sub>-. As a matter of convention, and for purposes of this specification

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involving hydrogenated polybutadiene: the notation -E- denotes at least about 85% of (-CH2-)4 units. The notation -B- denotes at least about 70% of [-CH2-CH(C2H5)-] units. The notation -EB- denotes between about 15 and 70% [-CH2-CH(C2H5)-] units. The notation -EBn- denotes n% [-CH2-CH(C2H5)-] units. For hydrogenated polyisoprene: The notation -EP- denotes about at least 90% [-CH2-CH(CH3)-CH2-CH2-] units"

and insert:

At page 11, lines 3, 5, 13, 17, and 21 correct the numbers "(0.5)4", "(0.6)4", "(0.25)4", "(0.10)4", and "(0.4)4" should be correct to read --- (0.5)4 ---, --- (0.6)4 ---, --- (0.25)4 ---,

--- (0.10)4 ---, and --- (0.4)4 --- respectively.

\*At page 11, lines 13 and 20, correct "CH2" to read --- CH2 ---.

At page 12, line 2, correct "(0.48)4" to read --- (0.48)4 ---.

At page 12, line 6, correct "Ebn" to read --- Ebn ---.

At page 12, lines 7-14, delete

"Table I below gives the % of units on hydrogenation of polybutadiene/polyisoprene copolymer midblocks

H2

-(CH2)4 -(CH-CH2) \_\_\_\_\_ (CH2-CH-CH2-CH2)-(CH2-CH)-

C2H5

CH3 CH

СНЗ СНЗ

n% from polybutadine

(1-n)% from polyisoprene

90%•n 10%•n

95%•(1-n) 5%•(1-n)

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where n is the mole % polybutadiene in the polybutadiene-polyisoprene starting polymer"

and insert:

--- Table I below gives the % of units on hydrogenation of polybutadiene/polyisoprene copolymer midblocks

 $(CH_2\text{-}CH\text{-}CH_2\text{-}CH_2)\text{-}(CH_2\text{-}CH)\text{-}$ 

CH<sub>3</sub>

CH<sub>3</sub> CH<sub>3</sub>

1045)

n% from polybutadine

(1-n)% from polyisoprene

90%•п

10%•n

95%•(1-n)

5%•(1-n)

where n is the mole % polybutadiene in the polybutadiene-polyisoprene starting polymer -- [...

At page 12, lines 22-24 and page 12, lines 1-7 delete:

"where R-1 denotes (-CH2-)4,

R-2 denotes -(CH-CH2)-,

C2H5

R-3 denotes -(CH2-CH2-CH2)-, and

CH3

R-4 denotes

-(CH2-CH)-

CH

СНЗ СНЗ



Therefore, the percentage that can crystallize is [(-CH2-)4]4 since this is the chance of getting four (-CH2-)4 units in sequence"

and insert:

--- where R-1 denotes (-CH<sub>2</sub>-)<sub>4</sub>,

R-2 denotes -(CH-CH<sub>2</sub>)-, C<sub>2</sub>H<sub>5</sub>

R-3 denotes  $-(CH_2-CH-CH_2-CH_2)$ , and  $CH_3$ 

C

R-4 denotes

-(CH<sub>2</sub>-CH)-CH CH<sub>3</sub> CH<sub>3</sub>

04/61)

Therefore, the percentage that can crystallize is [(-CH<sub>2</sub>-)<sub>4</sub>]<sub>4</sub> since this is the chance of getting four

(-CH<sub>2</sub>-)<sub>4</sub> units in sequence --

At page 13, correct the both table headings at lines 8 and 17:

4.0

n=	(-CH2-)4	[(-CH2-)4]4	0.6 X [(-CH2-)4]n
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to read ---

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n =	(-CH <sub>2</sub> -) <sub>4</sub>	[(-CH <sub>2</sub> -) <sub>4</sub> ] <sub>4</sub>	0.6 X [(-CH <sub>2</sub> -) <sub>4</sub> ] <sub>n</sub>
		· · · · · · · · · · · · · · · · · · ·	

-- respectively.

Х At page 13, line 15-16, correct "(-CH2-)4" to read --- (-CH<sub>2</sub>-)<sub>4</sub> ---.

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At page 16, line 25, 33, and 34 correct "(g) Kuraray SEPS 4033", "(29) Kuraray 4055 (S-EB-EP-S)", "(30) Kuraray 4077 (S-EB-EP-S)", and "(31) Kuraray 4045 (S-EB-EP-S)" to read:

- --- (g) Kuraray SEEPS 4033 ---, --- (29) Kuraray 4055 (S-E-EP-S) ---,
- --- (30) Kuraray 4077 (S-E-EP-S) ---, and --- (31) Kuraray 4045 (S-E-EP-S) --- respectively.

At page 18, lines 21-22 delete: "styrene to ethylene-ethylene/propylene (S:EB-EP) ratios of Kuraray's SEPTON 4045, 4055, and 4077 are" and insert: — styrene to ethylene-ethylene/propylene (S:E-EP) ratios of Kuraray's SEPTON 4045, 4055, and 4077 are — in-

At page 25, lines 1, 10, 16, 17 correct the exponents and dimensions "(5806 X 103)", "8X105 dyne/cm2", "8X105 dyne/cm2", "104 dyne/cm2", and at line 18 the exponents and dimensions "106 dyne/cm2", "104 dyne/cm2", "106 dyne/cm2" to read --- (5806 X 103) ---, --- 8X105 dyne/cm2 ---, --- 8X105 dyne/cm2 ---, and at line 18 the exponents and dimensions --- 106 dyne/cm2 ---, --- 104 dyne/cm2 ---, respectively.

At page 29, after line 3, insert (from page 20-25 of USSN 08/719,817, filed 9/30/96)

the following:

--- In order for gels to be useful as a dental floss, it must overcome the difficult barriers of high shearing and high tearing under extreme elongation and tension loads. The difficulties that the gels must overcome during flossing can be viewed as follows: during the action of flossing, the gel is stretched from no less than about 200% to about 1,100% or higher, the gel floss is deformed as it is pulled down with tearing action between the contacting surfaces of the teeth, then, the wedge of gel floss is sheared between the inner contacting surfaces of the teeth, and finally, the elongated wedged of gel floss is pulled upwards and out between the surfaces of the teeth. The forces encountered in the act of flossing are: tension, shearing, tearing under extreme tension.

This invention advances the flossing art by providing strong, soft, and extreme tear resistant gels made from multiblock copolymers which gels are substantially as soft as the gums surrounding the teeth.

Gel floss formed from the gels has many advantages over conventional dental floss such as regular and extra fine waxed and unwaxed nylon floss, spongy nylon fiber floss, and waxed and unwaxed expanded and unexpended teflon floss. Such conventional floss are not recommended for use by children, since a slip or sudden snap in forcing the floss between the teeth may cause injury to the gums which often times results in bleeding. For sensitive gums and inflamed gums which has become red and puffy, it is

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difficult to floss at, near, and below the gumline. The soft gel floss with softness substantially matching the softness of the gums are of great advantage for use by children and for flossing teeth surrounded by sensitive and tender gums.

The shear resistant characteristics of the gels can be indirectly determined by subjecting the gel to the shear forces of a pair of twisting strings and the resulting inward pulling forces of the twisting strings can be directly read off of a spring scale. As a pair of strings are gradually twisted, typical values will range from less than one pound to fifty pounds and greater. As the string is being twisted (simulating increased shearing forces), the measured pulling forces can range from a low value of 0.5, 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 20, 21, 22, 23, 24, 25, 26, 27, 28, 29, 30, 31.... to values of 40, 50, 60, 70, 80 pounds and greater.

Gel material of low strength can not resist the tremendous shearing action of the twisting strings. The twisting action of the strings can exhibit a first order twist, a second order twist, or higher order twists. A first order twist refers to one or more twists of a pair of strings (i.e. a pair of strings when twisted together forms a small tight binding helix). A second order twist refers to one or more large binding helixes build up by a pair of strings that have been twisted beyond the maximum number of twist which normally produce small tight binding helixes of the first order kind. Similarly, a third order twist refers to a much larger tightly binding helix build up by the maximum number of second order twists produced by the pair of twisting strings. The third order twist may be manifested by the appearance of a branch of two or more twist of the first order twisting strings.

The order of twisting will increase (from a one, two, three, and higher order twist) until the rubber band breaks. Likewise, a looped string with one end attached to a spring scale and the other end attached to a fixed anchor can be twisted into a first, second, third, and higher ordered twist state. This method can be utilized to directly measure the force generated for each ordered twist states. The static force generated by twisting a string on a spring scale is a way of determining the shear force generated in the shearing action of forcing the gel floss between two closely contacting teeth when flossing.

In considering dental flossing criteria, one or more of the following conditions can be regarded as critical factors for dental flossing gels.

#### SHEAR RESISTANT CRITERIA

For the gels to be considered useful for flossing, the gels, critically, should withstand a twisting string shearing force of at least about 5 Kg, more advantageously at least about 8 Kg, and still more advantageously at least about 10 Kg of inward pulling force of a pair of twisting strings measured directly on a spring scale.

### FLOSSING CYCLE CRITERIA

For the gels to be considered useful for flossing, the gels, critically, should advantageously be able to perform at least 4 flossing cycles, more advantageously 8 cycles, and still more advantageously of about

20 cycles without breaking apart when a 3.0 mm diameter gel strand is tested on a set of simulated upper front teeth fully contacting under a uniform spring load of (0.9027 Kg) two pounds. The simulated upper front teeth comprises two small stainless steel rollers (3/8" dia.) facing lengthwise parallel and forced together so as to form a contact length of 1/2 inches under a spring load of two pounds as measured by a Entran® model ELO-200-4 load cell adjusted by a straight micrometer at room temperature.

### **GEL STRENGTH CRITERIA**

For the gels to be considered useful for flossing, the gels, critically, should advantageously exhibit a tensile strength of at least 5 Kg/cm<sup>2</sup> (when extended to break as measured at 180° U bend around a 5.0 mm mandrel attached to a spring scale) and more advantageously at least 8 Kg/cm<sup>2</sup>, and still more advantageously of about 10 Kg/cm<sup>2</sup> and higher. The high and gels useful as dental floss can exhibit tensile strengths at break of at least 20 Kg/cm<sup>2</sup>, more advantageously of at least 40 Kg/cm<sup>2</sup>, and exceptionally more advantageously at least 60 Kg/cm<sup>2</sup>. Typically, the tensile strengths range from about 20 Kg/cm<sup>2</sup> to about 110 Kg/cm<sup>2</sup> and higher, more typically from about 30 Kg/cm<sup>2</sup> to 80 Kg/cm<sup>2</sup> and higher, and exceptionally typically from about 50 Kg/cm<sup>2</sup> to about 100 Kg/cm<sup>2</sup> and higher.

# PROPAGATING TEAR CRITERIA

As a minimum, for the Gels to be considered useful for flossing, the gels, critically, should advantageously exhibit a propagating tear force (when propagating a tear as measured at 180° U bend around a 5.0 mm diameter mandrel attached to a spring scale) of at least about 1 Kg/cm, more advantageously at least 2 Kg/cm, and still more advantageously of about 3 Kg/cm and higher. The gels useful as dental floss can exhibit tear strengths of at least 4 Kg/cm and higher, more advantageously of at least 6 Kg/cm and higher, exceptionally more advantageously of at least 8 Kg/cm and higher. Typically, the tear propagation strength can range from about 5 Kg/cm to about 20 Kg/cm and higher, more typically from about less than 5 Kg/cm to about 25 Kg/cm and higher, especially more typically form about less than 6 Kg/cm to about 30 Kg/cm and higher, and exceptionally more typically from about less than 8 Kg/cm to about 35 Kg/cm and higher.

For the Gels to be considered useful for flossing, the gels, critically, should advantageously exhibit a propagating tension tear force (when a cylindrical sample is notched and a tear is initiated at the notched area and propagated past its maximum cylindrical diameter by length-wise stretching of the cylindrical sample) of at least about 1 Kg/cm, more advantageously at least 2 Kg/cm, and still more advantageously of about 4 Kg/cm and higher. The extreme tear resistant gels typically will exhibit even higher tension tear values.

#### RIGIDITY CRITERIA

The rigidities of the extreme tear resistant useful for flossing can advantageously range from about



350 gram to about 1,800 gram Bloom, more advantageously from about 400 gram to about 1,500 gram Bloom, especially more advantageously from about 450 gram to about 1,200 gram Bloom, still more advantageously from about 450 gram to about 1,000 gram Bloom, and less advantageously at values of greater than 1,800 gram Bloom.

In general, as a minimum, the flossing gels should exhibit several critical properties, including advantageously the ability to:

- (1) withstand a shearing force of at least about 5 Kg under the string twisting test described above,
- (2) perform at least 4 flossing cycles without breaking apart when tested on a set of simulated upper front teeth fully contacting under a uniform spring load of two pound,
  - (3) exhibit a tensile strength of at least 5 Kg/cm<sup>2</sup> and higher,
  - (4) exhibit a propagating tear force at 180° U bend tear test of at least about 1 Kg/cm, and
- (5) exhibit a propagating tension tear force (on a notched cylindrical sample) of at least about 1 Kg/cm.

For use as a dental floss, the gel is made (by extruding, spinning, casting, etc) as a continuous gel strand, the gel strand can be in the shape of a fiber of a selected diameter (from less than about 0.15 to about 5.0 mm and greater) as a continuous tape having a selected width and thickness (less than 0.10 mm thin to about 5.0 mm and thicker) or in any desired shape suitable for flossing. The fiber, tape or a selected shape is then cut to a desired length, rolled up and placed into a dispenser suitable for containing and dispensing a measured use amount of gel floss. The continuous fiber and tape can be partly cut or notched for measured single or multiple use. When the floss is pulled from the dispenser to a point showing the notched or cut mark on the length of gel floss, the lid is pushed down on the gel floss nipping it and allowing the floss to be further pulled and separated at the notched or cut point. Additionally, a suitable floss dispenser containing a measured length of gel floss can be fitted with a cutting edge attached to its lid or on its body and the uncut and un-notched gel floss can be dispensed from the dispensing container and cut at the desired measured use length by pressing close the dispenser cutting edge down on the floss so as to nip and cut the gel or by simply closing the dispenser lid or running the gel along the cutting edge on the dispenser body separating a useful length of gel floss.

In practice, typically during flossing, a gel strand will under go various deformations, some of these deformations can be measured, including original shape, extended shape under tension, nipping force, and nipped deformation under a measured force and width. Typically, any shaped gel strand can be used for flossing, a square cross-section, a circular cross-section, a rectangular cross-section, round, oval, etc. For example, a 2.35 mm diameter strand when extended under a force of 2.5 kg can be nipped down to 0.14 mm thickness (along a 3 mm uniform width of its cross-section) by a force of 0.9072 Kg (2.0 pound



force), a reduction of 16.78:1; a 1.89 mm diameter strand when extended under a force of 2.5 kg can be nipped down to 0.14 mm thickness by a force of 0.9072 Kg (2.0 pound force), a reduction of 13.5:1; a 2.75 mm diameter strand when extended under a force of 2.5 kg can be nipped down to 0.19 mm thickness by a force of 0.9072 Kg (2.0 pound force), a reduction of 14.4:1; and a 2.63 mm diameter strand when extended under a force of 2.5 kg can be nipped down to 0.19 mm thickness by a force of 0.9072 Kg (2.0 pound force), a reduction of 13.8:1. the cross-section of the gel floss can be reduced to any degree by stretching and nipping (from less than about 1% to about 1,600% and higher). Advantageously, a gel having the required strength, tear resistance, gel rigidity, and other characteristics described can be formed into a floss of any selected cross-section and thickness provided the floss is capable of being stretched when flossing under tension without breaking. Typically the stretching or pulling force is from about less than 0.1 Kg to about 3 Kg and higher. The cross-section of the strand of gel floss should be capable of being nipped by a 0.9027 Kg (2 pounds) force applied across a width of 3 mm from its original cross-sectional dimensions to a nipped thickness of about 3.0 mm to about 0.02 mm and lower, more advantageously from about 2.5 mm to about 0.04 mm and lower, still more advantageously from about 2.0 mm to about 0.08 mm and lower; especially advantageously from about 1.5 mm to about 0.15 mm and lower; especially more advantageously from about 1.2 mm to about 0.20 mm and lower; especially still more advantageously from about 1.0 mm to about 0.25 mm and lower.

The gels made from higher viscosity copolymers (I) are resistant to breaking when sheared than triblock copolymer gels. This can be demonstrated by forming a very soft gel, for example 100 parts copolymer to 800 parts plasticizing oil. The soft gel is cut into a strip of 2,5 cm X 2.5 cm cross-section. the gel strip is gripped lengthwise tightly in the left hand about its cross-section and an exposed part of the gel strip being gripped lengthwise around its cross-section tightly by the right hand as close to the left hand as possible without stretching. With the two hands gripping the gel strip's cross-section, the hands are moved in opposite directions to shear apart the gel strip at its cross-section. The shearing action by the gripping hands is done at the fastest speed possible as can be performed by human hands. The shearing action is performed at a fraction of a second, possible at about 0.5 seconds. Using this demonstration, the copolymer (I) gels will not easily break completely apart as would gels formed from triblock copolymers. In some cases, it will take two, three, or more attempts to shear a high viscosity copolymer (I) gel strip this way. Whereas, a lower viscosity triblock copolymer gel strip can be sheared apart on the first try. For gels made from copolymers with viscosities of 5 wt% solution in Toluene, their shear resistance will decrease with decreasing viscosity. For example, the shear strengths as tested by hand shearing described above of gels made from copolymers having viscosities of 150, 120, 110, 105, 95, 90, 89, 85, 70, 60, 58, 48, 42, 40, 35, 28, 27, 25, 21 cps, and the like can be expected to decrease with decreasing viscosity.

The tensile strengths of multiblock copolymer gels made from higher viscosity copolymers (I) can be slightly lower than or equal to the tensile strengths of gels made from lower solution viscosity triblock

copolymers (II).

Strands of gels comprising higher viscosity multiblock copolymers will perform better than gel strands made from gels of lower viscosity triblock copolymers when used in flossing amalgam molars and more than three times better when used in flossing front teeth.

Gels, in general, will exhibit higher tensile and greater tear resistance than their parent gels containing higher concentrations of plasticizer.

As compared to spongy nylon, regular waxed nylon, and extra fine unwaxed nylon when flossing amalgam molars, the performance of multiblock copolymer gels are on the average substantially better.--

At page 33, after line 2, please insert the following:

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#### **EXAMPLE XV**

Gels of 100 parts of Kraton G1651, Kuraray Septon 2006 (SEPS), Kuraray Septon 8006 (SEBS), a high viscosity (SEB)<sub>n</sub>, and a high viscosity (SEP)<sub>n</sub> triblock copolymers and 1,600, 1,200, 1,000, 800, 600, 500, 450, and 300 parts by weight of Duraprime 200 white oil are melt blended and samples extruded (from a 7.15 mm diameter orifice) into selected lengths of varying diameters for use as dental floss, the bulk gel rigidities is found to be within the range of 2 to 1,800 gram Bloom, the tensile strength is found to decrease with increase orientation, and the optimum tensile strength found for gel samples with the least amount of stress or orientation imparted during cool from the molten state to room temperature.

#### **EXAMPLE XVI**

Example XV is repeated using Kuraray (S-E-EP-S) 4055 and 4077 multiblock copolymers, the bulk gel rigidities are found to be within the range of 2 gram to 1,800 gram Bloom and the tear resistance of the multiblock copolymers at corresponding rigidities are found to be substantially higher than the tear resistance of the triblock copolymer gels of EXAMPLE XV. The tensile strength is found to decrease with increase orientation, and the optimum tensile strength found for gel samples with the least amount of stress or orientation imparted during cool from the molten state to room temperature.

#### **EXAMPLE XVII**

Example XV is repeated using (S-E-EP-S), (S-E-EP-E-S), (S-B-EP-S), (S-E-EB-S), (S-E-EB-S), (S-E-EB-S), (S-E-EB-S), (S-E-EB-S), (S-B-EB-E-S), (S-B-EB-B-S), (S-B-EB-B-S), (S-E-E-EB-S), (S-E-E-EB-S), (S-B-E-EB-S), (S-B-E-EB-S), (S-B-E-EB-S), (S-B-E-EB-S), (S-E-E-EB-S), (S-E-EB-EB-S), (S-E-EB-EB-EB-S), (S-E-E





FROM :

B-EP-E-EP-S), (S-E-EB-B-EP-S), (S-E-EP-B-EB-S), (S-P-EB-S), (S-P-EP-S), (S-P-EP-S), (S-P-EB-P-S), (S-B-EP-P-S), (S-B-EB-P-S), (S-P-E-EP-S), (S-P-E-EB-S), (S-B-P-EP-S), (S-B-P-EB-S), (S-P-B-EB-S), (S-P-B-EP-S), (S-P-EB-EB-S), (S-P-EP-EP-S), (S-P-EB-EP-S), (S-P-EP-EB-S), (S-P-EP-P-EP-S), (S-P-EB-P-EB-S), (S-P-EP-P-EB-S), (S-B-EB-P-EB-S), (S-B-EP-P-EP-S), (S-P-EB-B-EP-S), (S-P-EP-B-EB-S), (S-E-EP-P-S), (S-E-EB-P-S), EP-S), (S-E-P-EB-S), (S-E-EP-P-EP-S), (S-E-EB-P-EB-S), (S-E-EP-P-EB-S), (S-E-EP-E-EP-E-S), (S-B-EP-B-EP-B-S), (S-P-EP-P-EP-P-S),  $(S-E-EB-E-EB-E-S), (S-P-EP-P-EP-P-S), (S-E-EP)_n, (S-E-EP-E)_n, (S-B-EP)_n$  $(S-E-EB-S)_n$ ,  $(S-EB-EP-)_n$ ,  $(S-E-EP-EB)_n$ ,  $(S-B-EB)_n$ ,  $(S-E-EB-E)_n$ ,  $(S-B-EP-E)_n$  $(S-B-EB-E)_n$ ,  $(S-B-EP-B)_n$ ,  $(S-B-EB-B)_n$ ,  $(S-E-E-EP)_n$ ,  $(S-E-E-EB)_n$ ,  $(S-B-E-EP)_n$  $(S-B-E-EB)_n$ ,  $(S-B-B-EP)_n$ ,  $(S-B-B-EB)_n$ ,  $(S-E-B-EB)_n$ ,  $(S-E-B-EP)_n$ ,  $(S-E-E-EB)_n$ ,  $(S-EP-EP)_n$ ,  $(S-E-EB-EB)_n$ ,  $(S-E-EP-EP)_n$ ,  $(S-E-EB-EP)_n$ ,  $(S-B-EB-EB)_n$ ,  $(S-B-EP-EP)_n$ ,  $(S-B-EB-EB)_n$ EP)<sub>n</sub>, (S-B-EP-EB)<sub>n</sub>, (S-E-EP-E-EP)<sub>n</sub>, (S-E-EB-E-EB)<sub>n</sub>, (S-E-EP-E-EB)<sub>n</sub>  $(S-B-EP-B-EP)_n$ ,  $(S-B-EB-B-EB)_n$ ,  $(S-B-EB-B-EP)_n$ ,  $(S-B-EB-E-EB)_n$ ,  $(S-B-EP-E-EP)_n$  $(S-E-EB-B-EP)_n$ ,  $(S-E-EP-B-EB)_n$ ,  $(S-P-EB)_n$ ,  $(S-P-EP)_n$ ,  $(S-P-EP-P)_n$ ,  $(S-P-EB-P)_n$ ,  $(S-B-EP-P)_n$ ,  $(S-B-EB-P)_n$ ,  $(S-P-E-EP)_n$ ,  $(S-P-E-EB)_n$ ,  $(S-B-P-EP)_n$ ,  $(S-B-P-EB)_n$ ,  $(S-P-B-EB)_n$ ,  $(S-P-B-EP)_n$ ,  $(S-P-EB-EB)_n$ ,  $(S-P-EP-EP)_n$ ,  $(S-P-EB-EP)_n$ ,  $(S-P-EP-EB)_n$ ,  $(S-P-EP-EP)_n$ EP)<sub>n</sub>, (S-P-EB-P-EB)<sub>n</sub>, (S-P-EP-P-EB)<sub>n</sub>, (S-B-EB-P-EB)<sub>n</sub>, (S-B-EP-P-EP)<sub>n</sub>,  $EP_{n}$ ,  $(S-E-EB-P-EB)_{n}$ ,  $(S-E-EP-E-EB)_{n}$ ,  $(S-E-EP-E-E-E)_{n}$ ,  $(S-B-EP-B-EP-B)_{n}$ ,  $(S-P-EP-P-EP-P)_{n}$ (S-E-EB-E-EB-E)<sub>n</sub>, and (S-P-EP-P-EP-P)<sub>n</sub> multiblock copolymers, the bulk gel rigidities are found to be within the range of 2 gram to 1,800 gram Bloom and the tear resistance of the multiblock copolymers at corresponding rigidities are found to be substantially higher than the tear resistance of the triblock copolymer gels of EXAMPLE XV. The tensile strength is found to decrease with increase orientation, and the optimum tensile strength found for gel samples with the least amount of stress or orientation imparted during cool from the molten state to room temperature.

#### **EXAMPLE XVIII**

Example XVI is repeated using plasticizers L-14, L-50, L-100, H-15, H-25, H-35, H-50, H-100, H-300, L-14E, H-300E, Actipol E6, E16, E23, Kraton L-1203, EKP-206, EKP-207, HPVM-2203, Amoco C-60, Piccolyte S10, Duraprime (55, 70, 90, 200, 350, 400), Tufflo (6006, 6016, 6016M, 6026, 6036, 6056, 6206,) Bayol, Bernol, American, Blandol, Drakeol, Ervol, Gloria, and Kaydol, the bulk gel

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rigidities are found to be within the range of 2 gram to 1,800 gram Bloom and the tear resistance of the multiblock copolymers at corresponding rigidities are found to be substantially higher than the tear resistance of the triblock copolymer gels of EXAMPLE XV.

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#### **EXAMPLE XIX**

Example XVII is repeated using plasticizers L-14, L-50, L-100, H-15, H-25, H-35, H-50, H-100, H-300, L-14E, H-300E, Actipol E6, E16, E23, Kraton L-1203, EKP-206, EKP-207, HPVM-2203, Amoco C-60, Piccolyte S10, Duraprime (55, 70, 90, 200, 350, 400), Tufflo (6006, 6016, 6016M, 6026, 6036, 6056, 6206,) Bayol, Bernol, American, Blandol, Drakeol, Ervol, Gloria, and Kaydol, the bulk gel rigidities are found to be within the range of 2 gram to 1,800 gram Bloom and the tear resistance of the multiblock copolymers at corresponding rigidities are found to be substantially higher than the tear resistance of the triblock copolymer gels of EXAMPLE XV.

#### **EXAMPLE XX**

A gel composition of 100 parts of Kuraray S-E-EP-S 4055 copolymer and 400 parts by weight of Duraprime 200 white oil was made following Example XV and extruded and drawn (from a 7.15 mm diameter orifice) into a strand of uniform diameter onto a take-up roll of continuous lengths. The strand diameter was varied by increasing and decreasing the speed of the take-up roll. The continuous strand of varying diameter gel strand was cut to suitable lengths for use and testing as dental floss. Additional gel was also casted in varying thickness and tested. The results of samples tested are shown in Table 3, #4-7; Table 4, #12-15 and 20; Table 5 #22, 23, 27-29; Table 6 #36-32; Table 7, #40-43, #76 and 77. Sample Nos. 76 and 77 were tested together. Sample 77 exhibited higher tensile strength after 27.75% of plasticizing oil was extracted (with 2.89 parts by weight of oil remaining), its rigidity remained substantially unchanged.

#### **EXAMPLE XI**

A gel composition of 100 parts of Kraton G1651 and 400 parts by weight of Duraprime 200 white oil was made following Example XV and extruded and drawn (from a 7.15 mm diameter orifice) into a strand of uniform diameter onto a take-up roll of continuous lengths. The strand diameter was varied by increasing and decreasing the speed of the take-up roll. The continuous strand of varying diameter gel strand was cut to suitable lengths for use and testing as dental floss. Additional gel was also casted in varying thickness and tested. The results of samples tested are shown in Table 3B, #8-11; Table 4, #16-19 and 21; Table 5, #24-26; Table 6, #33-35; and Table 7, #36-39.





## **EXAMPLE XXII**

Example XVI was repeated melt blending under inert gas 100 parts by weight of Kuraray (S-E-EP-S) 4077 multiblock copolymer and 400 parts by weight of Duraprime 70 white oil. A first part of the molten gel was allowed to cool to room temperature, the remainder gel was heated under inert gas for an additional three hours at 300-3250F and a second part of the gel was extruded (from a 7.15 mm diameter orifice) into cold running water, and the third and final remaining gel was allowed to cool to room temperature. The bulk gel rigidities of the first, second and third parts were found to be within the range of 2 to 1,800 gram Bloom. The second and third final parts of the gel appeared to be altered and different from the first gel part. The first part exhibited rapid return when extended, but the second and third final parts exhibited delay elastomeric recovery when released after extension and deformation. All of the samples exhibited 100% recovery after repeated extensions and deformations.

Table 3A. Flossing Cycles to Break

Sample No.	Floss Type	cross-section size	<sup>2</sup> Floss amalgam molars to break	<sup>3</sup> Floss fronts to
1	4Unwaxed spongy nylon	0.30	18	200+
2	5Regular waxed nylon	0.11	11	200+
3	<sup>6</sup> Extra fine unwaxed nylon	0.06	6	200+

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# Table 3B. Flossing Cycles to Break

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Sample No.	Floss Type	<sup>1</sup> Relaxed/extended dia. (mm)	<sup>2</sup> Floss amalgam molars to break	3Floss fronts to break
4	<sup>7</sup> Gel	2.42/0.16	37	76
5	<sup>7</sup> Gel	2.63/0.17	29	83
6	7Gel	2.75/0.17	36	183
7	7Gel	2.83/0.20	20	74
8	8Gel	3.22/0.22	8	30
9	8Gel	2.48/0.31	4	20
10	8Gel	3.16/0.33	6	44
11	8Gel	2.86/0.24	5	29

Ifloss dimension relaxed state and when extended during flossing cycles. 2Test conditions: number of flossing cycles (before breaking) between amalgam alloy metal (fully contacting) lower, left first and second human back molars. 3Test conditions: number of flossing cycles (before breaking) between upper human front teeth. 4Oral-B Ultra Floss™, interlocking network of spongy nylon floss. 5Johnson & Johnson regular waxed nylon floss. 6Johnson & Johnson extra fine unwaxed nylon floss. 7Gel made from 100 parts by weight of S-E-EP-S 4055 multiblock copolymer having a Brookfield viscosity of 90 as measured for a 5wt% solution in toluene at 30°C and 400 parts by weight of Duraprime 200 plasticizing oil. 8Gel made from 100 parts by weight of SEBS Kraton G1651 copolymer having a Brookfield viscosity of 40 as measured for a 5wt% solution in toluene at 30°C. 2.3 Any selected test methods may be utilized in testing the floss performance of the gels. For example, a set of simulated upper front teeth fully contacting under a uniform spring load of two pounds may be used in place of human teeth. Such simulated testing conditions may be more severe than conditions 2 and less severe than conditions 3 above.



Table 4. Tensile Strength of Gel Strands

Sample No.	Number of Strands	Radius (mm)	Area (cm <sup>2</sup> )	Failure (Kg)	Tensile (Kg/cm <sup>2</sup> )
12	3	1,325	0.165	9.00	54.54
13	4	1.250	0.196	9.50	48.39
14	4	1.421	0.253	9.50	37.44
15	5	1.359	0.290	12.5	43.08
16	2	2.14	0.287	14.0	48.78
17	2	1.55	0.151	11.5	75.95
18	2	1.17	0.086	8.50	98.84
19	2	1.322	0.109	9.0	81.96
20	6	1.375	0.356	14	39.32
21	2	1.445	0.131	10	76.33
76	1	1.22	0.0467	2.00	42.82
77†	1	1.38	0.0598	4.00	66.88

†Plasticizing oil extracted



Table 5. Tensile Strength of Bulk Gels Samples

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Sample No.	Cross-section	Failure	Tensile
No	(cm2)	(Kg)	(Kg/cm2)
22	1.96	24.0	12.24
23	1.56	25.0	16.02
24	0.58	15.0	25.83
25	0.602	16.0	26.54
26	1.163	24.0	20.64
27	0.913	21.0	23.00
28	0.595	18.5	36.56
29	0.702	19.0	27.06

Table 6. 180 U Bend Tear Propagation of Bulk Gels Samples

Sample No.	Tear width (cm)	Failure (Kg)	Tear Force (Kg/cm)
30	1.31	2.75	2.09
31	1.28	3.0	2.30
32	1.14	2.75	2.56
33	1.53	2.75	1.79
34	1.27	2.25	1.76
35	1.26	2.25	1.77

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# Table 7. Notched Gel Strand Tension Tear Propagation

Sample No.	Strand Dia. (mm)	Failure (Kg)	Tear Force (Kg/cm)
36	2.86	0.75	2.62
37	2.49	0.75	3.01
38	3.09	0.60	1.94
39	2.62	0.70	2.67
40	2.54	0.60	2,36
41	1.94	1.10	5.67
42	1.58	0.75	4.74
43	2.34	1.2	5.12

The tensile strengths of gels made from higher viscosity copolymers are lower than the tensile strengths of gels made from lower solution viscosity copolymers. This was later found to be due to orientation effects and not considered significant.

The tear resistance of gels made from higher viscosity copolymers are higher than the tear resistance of gels made from lower solution viscosity copolymers.

Gel strands made from higher viscosity copolymers perform better than gel strands made of lower viscosity copolymers when used in flossing amalgam molars and more than three times better when used in flossing front teeth.

As compared to spongy nylon, regular waxed nylon, and extra fine unwaxed nylon when flossing amalgam molars, the performance of gels are on the average substantially better.

Examples below illustrate other modes of practice contemplated.

# **EXAMPLE XXIII**

At least 120 PCs of the gel strands of EXAMPLE XVI containing 600 parts oil is individually weighted and placed in a heated vacuum oven, a partial vacuum is applied and the temperature is regulated between about 80°F to about 150°F to extract plasticizer from the gel strands. At various oven and vacuum times, three gel strands are removed from the vacuum oven, allowed to cool to room



temperature, weighted to determine the amount of weight loss and tested for tensile and tear strength. As the amount of oil contained in the original gel is reduced from 600 parts by weight to less than 200 parts by weight, the "reduced plasticizer volume" gels are weighted and tested. The tear and tensile strengths of the reduced plasticizer volume gels are found to be improved over the properties of the original 600 parts by weight referenced gel strands.

The gels are especially advantageously useful when subjected to conditions of stretching, shearing, and tearing during flossing. The gels useful for flossing are characterized by low rigidities and high solution viscosity of the gels made from multiblock copolymers having two or more midblock polymer chains.

Tables 8-11 are illustrative in meeting one or more of the criteria detailed above.

# 8. Illustrative Modes of Practice Contemplated for multiblock copolymer Gels

100 Parts by wt	5Wt% Copolymer Viscosity (cps)	Styrene %	Parts by Wt of Oil	Number of floss cycles to break	Sample No.
S-E-EP-S	90	30	300	30+	44
S-E-EP-E-S	60	30	300	30+	45
(S-E-EP)n	240	35	300	30+	46
(S-E-EP-E)n	240	35	300	30+	47
S-B-EP-S	90	30	300	30+	48
S-E-EB-S	90	35	300	30+	49
S-EB-EP-S	90	30	300	30+	50
S-E-EP-EP-S	90	30	300	30+	51

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Table 9. Illustrative Modes of Practice Contemplated for multiblock copolymer Gels

100 Parts by wt	SWt% Copolymer Viscosity (cps)	Styrene %	Parts by Wt of Oil	Number Floss cycles to Break	Sample No.
S-E-EP-EB-S	120	33	250	30+	52
S-E-EP-EP-S	120	33	250	30+	53
(S-B-EP)n	380	35	250	30+	54
(S-E-EB)n	380	35	250	30+	55
S-E-EP-E-EP-S	120	30	250	30+	56
S-E-EP-P-S	120	35	250	30+	57
S-E-B-EP-S	120	30	250	30+	58
S-E-EP-EP-E-S	120	30	250	30+	59

Table 10. Illustrative Modes of Practice Contemplated for multiblock copolymer (0.5 - 2.0 cm diameters) Gel Strands

100 Parts by wt	5Wt%	Styrene	Parts by Wt	# Floss cycles to	Sample
	Copolymer	%	of Oil	Break	No.
	Viscosity (cps)	1			
S-E-EP-S	40	30	350	30+	60
S-E-EP-S	60	30	350	30+	61
(S-E-EP-EB)n	340	30	350	30+	62
(S-E-EP-EP-E)n	340	30	350	30+	63
S-E-EP-E-EP-E-S	90	30	350	30+	64
S-EB-EP-EP-S	90	35	350	30+	65
S-B-EB-B-\$	90	30	350	30+	66
S-E-EP-EP-E-S	90	30	350	30+	67
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Table 11. Illustrative Modes of Practice Contemplated for multiblock copolymer (0.5 - 2.0 cm diameters) Gel Strands

100 Parts by wt	5Wt% Copolymer Viscosity (cps)	Styrene %	Parts by Wt of Oil	# Floss cycles to Break	Sample No.
S-E-EB-S	120	30	250	40+	68
S-E-EP-S	120	30	250	40+	69
(S-E-EB)n	280	35	250	40+	70
(S-E-EP)n	280	35	250	40+	71
S-E-EP-E-S	120	30	250	40+	72
S-EP-E-EP-S	120	30	250	40+	73
S-EB-E-EB-S	120	30	250	40+	74
S-EB-EB-S	120	30	250	40+	75

#### REMARKS

The application and the material cited to date have been carefully reviewed along with Examiner's remarks in the Office action. After this review, Applicant is convinced that his claimed composition and articles are novel and patentable. Applicant strongly believes that his application embodies a significant discovery, that the claims define the invention in a clear and definite manner, and that all of the amended claims are allowable.

Claims 1-14 are pending in the case.

The amendments of pages 1-25 are directed to correction of transcription errors. The amendment of pages 29 and 33 do not involve new matter and are supported by the application as original filed incorporated by reference from USSN: 08/719,817; 08/863,794; and 09/230,940 which the present application is a C-I-P. These applications are directed to crystalline or crystal gels like the present claimed

